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Soot Particle Optical Properties: a Comparison between Numerical Calculations and Experimental Data Collected during the Boston College Experiment

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The Boston College experiment was conducted in July 2008 to carry out an inter-comparison study of various instruments used to measure the optical, physical and chemical properties of laboratory generated soot under controlled conditions (Cross, Onasch et al. 2010). The physical, chemical and optical properties were measured on size-selected particles for: 1. Nascent soot particles 2. Nascent- denuded soot particles 3. Soot particles coated with sulfuric acid and DOS (dioctyl sebacate) for different coating thicknesses 4. Coated and then denuded soot particles. Instruments involved in the inter-comparison study fell into two broad categories: a) massbased instruments and b) optically-based instruments. Mass-based instruments measured mass related properties of aerosol particles whereas optically-based instruments measured optical properties like scattering, absorption and extinction at different wavelengths. During the Boston College experiment, 7 mass-based and 9 optically-based instruments were deployed. Absorption scattering and extinction measurements were carried out in combination with mass-based instruments in order to obtain absorption scattering and extinction coefficients for coated and denuded soot particles as a function of their mass, size and coating thickness. In particular, a 3 laser integrated photoacoustic and nephelometer spectrometer was deployed during the experiment. The instrument measured aerosols at 405, 532 and 780 nm providing wavelength-dependent extensive (absorption and scattering) and intensive (single scatter albedo and Ångstrom exponents) aerosol optical properties. Particle samples were also collected on nuclepore filters to perform Scanning Electron Microscopy (SEM) analysis. The images obtained with the SEM elucidated the changes in particle morphology with particle generation process, coating and denuding. The images were also used to determine morphological parameters for single soot aggregates (e.g. monomers number and diameter) used in the numerical estimation of aerosol optical properties.

With the data collected during this experiment, we carry out a comparative study of the optical properties of soot particles obtained experimentally with those calculated using numerical calculations, to validate the degree of agreement between theoretical models and experimental results. The Rayleigh-Debye-Gans-approximation (RDG) and the Mie scattering techniques are used in this comparison. RDG approximation is a convenient approximation often used to model the optical properties of agglomerates, while the Mie scattering theory is rigorously valid only for particles with spherical symmetry, it is also often used to approximate

the optical properties of irregularly-shaped particles when morphology information are missing or to reduce computational times. Our goal is to use laboratory optical, mass, size and morphological data to study the impact of these parameters on radiative forcing by processed soot (Jacobson 2001; Flowers, Dubey et al. 2010).

References:

- Cross, E. S., T. B. Onasch, et al. "Soot Particle StudiesInstrument Inter-ComparisonProject Overview." Aerosol Science and Technology 44(8): 592-611.
- Flowers, B. A., M. K. Dubey, et al. (2010). "Correlations between aerosol optical properties and chemical composition in mixed carbonaceous aerosol: Observations on Jeju Island, Korea with a 3-laser photoacoustic spectrometer." Atmos. Chem. Phys. Under revision.

Jacobson, M. Z. (2001). "Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols." Nature 409(6821): 695-697.